

DECLARATION OF KOUICHI TAKEMOTO
UNDER 37 CFR §1.132

I am Kouichi Takemoto. I have worked in the field of electronic materials for ten years at Nippon Mining & Metals Co., Ltd. I am familiar with the properties of metals commonly used to manufacture semiconductor devices. In particular, I am skilled in the art of producing high purity copper.

I have read the publication titled "Ductility of Ultra High Purity copper" (Document D1). I have also read the specification of U.S. Patent No. 5,206,430 (Document D2). I confirm that I fully understand both of these documents.

According to the "8N-Cu" analysis (Table 1) disclosed in D1, included as impurities are Ag: 0.011 ppm, Al: 0.003 ppm, Si: 0.023 ppm, Ti: 0.004 ppm, and Fe: 0.002 ppm, and the total content of impurities is 0.043 ppm. Accordingly, when considering that the content of other impurities is not zero, the purity of "8N-Cu" disclosed in D1 is less than 99.9999957% (to 7N5), and fails to satisfy the purity requirement (8N or higher) of the present invention. Moreover, although D1 does not refer to the RRR of its "8N-Cu," the purity of its "8N-Cu" is less than 7N5. According to the relation (Table 1) of the purity and RRR disclosed in the present specification, it is speculated that the RRR of "8N-Cu" of D1 fails to satisfy the requirement (80,000 or more) of RRR of the present invention.

In addition, the ultra high purity copper of the present invention is not obvious to a person skilled in the art. The reasons for this are (1) a part of the purification process described in the present specification is not predictable by a person skilled in the art, and (2) the effect of the ultra high purity copper of the present invention is also not predictable by a person skilled in the art.

With respect to reason (1) above, D2 discloses a purification method based on activated carbon, but this method cannot simply be applied to the purification of the copper electrolyte of the present invention. The reasons for this are explained below. What is most difficult upon obtaining a purity level of 8N or higher from a purity level of less than 8N

is the elimination of Ag. Under normal circumstances, the elimination of Ag is performed by adding chlorine ions and precipitating/eliminating this as AgCl. Nevertheless, since only trace amounts of AgCl will precipitate when the purity level becomes approximately 8N, it cannot be separated with standard filtration. Thus, activated carbon is added for eliminating AgCl, and AgCl is adsorbed thereto and eliminated together with the activated carbon. Here, the solubility of AgCl has temperature dependency, and the solubility becomes lower as the temperature is lower (refer to Fig. 1). In cases where the purity level is 8N or higher, the difference in solubility will directly affect the purity level. Thus, in the series of processes of precipitating Ag as AgCl, adding activated carbon, adsorbing AgCl thereto, and filtering the same, it is vital to lower the temperature of the solution as much as possible. It is not possible to simply apply the purification method based on activated carbon without giving any consideration to the above.

Fig. 1 shows the solubility of silver relative to the solution temperature in the copper sulfate solution (copper concentration: 50 to 90 g/L). The average value and its standard deviation are shown with the pH of the electrolyte at 1, 1.5, and 2.

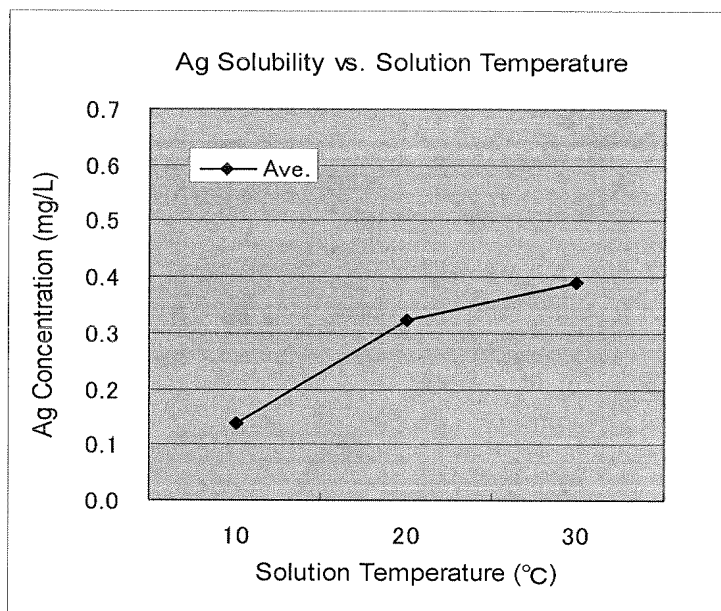


Fig. 1 Solubility of AgCl in Copper Sulfate Solution

In relation to reason (1) above, the present invention adopts a method of partitioning the anode and the cathode with an ion exchange membrane. Although this method is described in conventional technology (Japanese Patent Laid-Open Publication No. H11-229172), its object and effect are different from the present invention. The reason why H11-229172 partitions the anode and the cathode with an ion exchange membrane is “to leave elements forming the complex ions of platinum group metals and the like in the anode chamber” as described in paragraph [0011]. Meanwhile, the present invention partitions the anode and the cathode with an ion exchange membrane in order to precipitate the problematic Ag as AgCl by adding Cl ions, and thereby eliminating such AgCl with a micro filter. It is standard not to use a diaphragm in the electrolytic refining of copper, but in the case of the present invention, electrowinning technology (partitioning the anode and the cathode with an ion exchange membrane), which is able to reliably eliminate Ag, is employed in order to achieve a high purity level of 8N or higher. As described above, even if it is the same partitioning technology that uses an ion exchange membrane, so as long as the objective and effect of using the technology are different, the technology of the present invention and the technology of H11-229172 can no longer be viewed as the same technology.

With respect to reason (2) above, the effects of the ultra high purity copper of the present invention are, as shown in Table 1 of the specification, “Temperature Measurement Standard Sample: [Excellent]” and “Number of Particles: [Minimal],” and these effects could not have been predicted by a person skilled in the art.

The ultra high purity copper of the present invention has a higher purity level and RRR in comparison to the “8N-Cu” disclosed in D1. Consequently, the present invention yields extremely advantageous effects that could not have been predicted by a person skilled in the art. In addition, the method of producing the ultra high purity copper with a purity level of 8N or higher according to the present invention is not a method that could have been predicted by a person skilled in the art. The present invention was not obtained as a result of inventing the difference in purity levels.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like are punishable by fine and imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: 2nd / Nov. / 2010

KOICHI TAKEMOTO

Signature: Kouichi Takemoto